14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010

The Electronic and Magnetic Properties of $Yb_xGd_{1-x}Ni_5$ Systems

A. Szajek, M. Werwiński

Institute of Molecular Physics, Polish Academy of Sciences, M. Smoluchowskiego 17, 60-179 Poznań, Poland

G. Chełkowska, A. Bajorek and A. Chrobak

A. Chełkowski Institute of Physics, Silesian University, Uniwersytecka 4, 40-007 Katowice, Poland

The intermetallic compounds $Yb_xGd_{1-x}Ni_5$ crystallize in the hexagonal $CaCu_5$-type structure. Based on wide ranging SQUID-type magnetometer, it was shown that the saturation magnetization decreases with growing concentration of ytterbium. The opposite tendency was observed for the Sommerfeld coefficient obtained in the heat capacity measurements. These results are confirmed using *ab initio* band structure calculations.

PACS numbers: 71.20.−b, 75.25.−j

1. Introduction

The RNi_5 compounds (R = rare earth) have been intensively studied in recent decades [1–12]. Newly obtained series of the $Yb_xGd_{1-x}Ni_5$ compounds ($x \leq 0.5$) exhibit interesting properties [5]. All studied compounds crystallize in the hexagonal $CaCu_5$ type structure. However, the Yb/Gd substitution causes the increase in $c(x)$ lattice parameter in the whole range of studied concentrations. This kind of change is connected with different ionic radii for gadolinium and ytterbium ions. The deviation from the linear change of $a(x)$ and $V(x)$ for $x > 0.4$ may be related to an intermediate valence of Yb ions. The properties of the $Yb_xGd_{1-x}Ni_5$ compounds are very interesting and were the motivation to study their electronic and magnetic properties more in detail using *ab initio* calculations and SQUID measurements.

2. Experimental and computational details

Polycrystalline samples of the $Yb_xGd_{1-x}Ni_5$ series with $x = 0.0$, 0.2, 0.4, 0.5 were prepared by an arc melting technique from high purity elements under the argon atmosphere. The purity of materials used for preparing samples were 99.99% for Ni and Yb and 99.9% for Gd. The crystal structure of all samples was checked by means of X-ray diffraction (XRD) using Siemens D5000 diffractometer. The magnetic properties of $Yb_xGd_{1-x}Ni_5$ compounds were measured with the use of SQUID magnetometer (MPMS XL7 Quantum Design). All measurements were performed in the 2–400 K temperature range up to 7 T magnetic field.

In the band structure calculations we employed the full potential linear augmented plane wave (FP LAPW [13]) method in Wien2k code implemented in the latest version of the original Wien code. The calculations were performed within the density-functional theory for experimental lattice constants [5]. We started for $x = 0$ from the $CaCu_5$-type unit cell and for other concentrations appropriate supercells were constructed to get an experimental stoichiometry. We used the generalized gradient approximation (GGA) [14] to the potential parameterized in the form proposed by Perdew et al. [14]. The self-consistent criterion for the total energy was equal to at least 10^{-6} Ry.

3. Results and discussion

The isothermal magnetization curves $M(H)$ for the $Yb_xGd_{1-x}Ni_5$ system were measured at 2 K and up to 7 T. The magnetization saturates faster for the Gd-rich compounds. The highest value of the saturation magnetization M_S is observed for $x = 0.0$ and slowly decreases with the increase of ytterbium concentration up to $x = 0.5$ (see Table).The value of M_S for the $GdNi_5$ compound is smaller than for free Gd^{3+} ion (7 μ_B). As we suppose from X-ray photoemission spectroscopy (XPS) measurements [5] the ytterbium ions in the $Yb_xGd_{1-x}Ni_5$ series are visible in trivalent state. Moreover, magnetic Yb^{3+} ions have a smaller magnetic

TABLE

The electronic and magnetic characteristics for $Yb_xGd_{1-x}Ni_5$ series: saturation magnetization M_S and total magnetic moment m [μ_B/f.u.], measured (γ) and calculated (γ_0) Sommerfeld coefficient [mJ/(mol K^2)], density of states (DOS)at the Fermi level [states/(eV spin f.u.). In parenthesis there are collected calculated magnetic moments on Gd and Yb atoms.

x	M_S	γ	DOS(E_F) spin ↑ / ↓	γ_0	m
0.0	6.52	45.3	4.66/4.54	21.7	7.08 (Gd:6.88)
0.2	5.72	73.1	1.45/5.71	16.9	7.68 (Gd1:6.82/Gd2:6.82/Yb:0.80)
0.4	4.66	73.6	2.74/6.85	22.6	3.43 (Gd1:6.82/Gd2:-6.94/Yb:0.50)
0.5	4.36	104.2	9.23/1.83	26.1	0.96 (Gd:6.94/Yb:-0.81)

moment than Gd^{3+}. Therefore Yb/Gd substitution causes the gradual decrease of the $M_S(x)$ value.

The Sommerfeld coefficient γ was fitted from specific heat measurements $C(T) = \gamma T + C_D(T)$, where $C_D(T)$ is the Debye expression for the phonons, for $T > T_C$, and results are collected in Table. Their values increase with an increasing content of Yb atoms. For related compound $DyNi_5$ $\gamma = 36$ mJ/(mol K^2) [15].

In Fig. 1 we present DOS plots for $Yb_xGd_{1-x}Ni_5$ systems calculated based on Wien2k code within the GGA approach. It is our first attempt to this problem in spite of known limitations of GGA, which are manifested by wrong positions of $4f$ peaks. The calculated values of DOS at the Fermi level together with the Sommerfeld coefficients are collected in Table. Values of γ_0 are much lower than γ experimental, the difference indicates important role of many-body effects in these systems, which are described by the simple formula $\gamma = \gamma_0(1+\lambda)$, where λ is the mass enhancement factor.

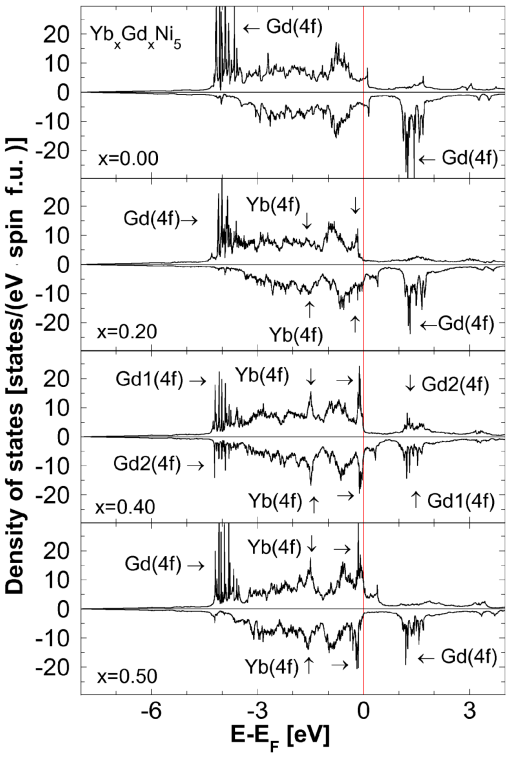

Fig. 1. Total and spin projected DOS plots for $Yb_xGd_{1-x}Ni_5$ systems ($x = 0.0$, 0.2, 0.4, and 0.5).

To get magnetic moments we started calculations from different initial moments assuming their anti- or parallel alignment, then the total energy was compared. Results presented in Fig. 1 and Table are for the case with the minimum energy. The general tendency in the calculated magnetic moment per formula unit and Sommerfeld coefficient is compatible with experimental results, but to improve the description of the strongly correlated $4f$ electrons, crucial role of many-body effects should be taken into account (see [16] for elemental, metallic Yb). The magnetic structure of considered systems is complicated, usually Ni atoms treated as non-magnetic are magnetic with moments up to 0.4 μ_B/atom and have different orientations.

4. Conclusions

Presented results may be summarized as follows:

— the substitution of Yb for Gd atoms up to $x = 0.5$ in the $GdNi_5$ crystalline compound causes the decrease of the saturation magnetization, from 6.52 μ_B/f.u. to 4.36 μ_B/f.u. for $x = 0$ to $x = 0.5$, respectively;

— the Sommerfeld coefficient increases with increasing content of Yb;

— above conclusions are confirmed by *ab initio* calculations.

Acknowledgments

This work was supported by MNiSW within grant No. NN202134933.

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